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The prompt γ-ray spectra from ^{235}U(n,f) at incident energies of 1 to 20 MeV and for ^{235}Cf(s.f.) were measured up to 4 MeV in coincidence with two parallel plate avalanche counters and the liquid scintillator array FIGARO. The unfolded γ-ray spectra from 1-4 MeV using the single value decomposition and iterative Bayesian techniques for the incident neutron energy regions corresponding to the first, second and third chance fissions were found to nearly identical and similar to the distribution from the spontaneous fission of ^{252}Cf. General agreement with the γ-ray distributions from fission was also found with previous measurements.

I. INTRODUCTION

Nuclear energy will become increasing important in the desire to reduce greenhouse gas emissions by 2050. In 2009, is was the second largest low-carbon emitting energy source producing 13.4% of the world's electricity [1]. Most of the available energy created from fission of a nucleus is carried off by the recoiling fragments in the form of kinetic energy of fragments, but $\sim$10% of the total energy in a core of a reactor is released in the form of prompt, delayed and radiative capture γ-rays [2]. Since γ rays may be the dominate contributor of the heating in the sub-assemblies and shielding of fast breeder reactors [3], detailed knowledge about the distributions of γ rays from fissile materials such as uranium and plutonium is needed for the designing of proper shielding and cooling systems. Information on observables from fission such as the kinetic energy of the emitted neutron and γ-rays are important to nuclear energy designs and safeguards scenarios. Accurate data on the properties of fission products is also needed to improve our understanding of the fission process and to test the accuracy of nuclear reaction codes. In this short manuscript, we will discuss our experimental efforts and progress using two unfolding techniques, Single Value Decomposition (SVD) and iterative Bayesian, to examine one of the least studied quantities of fission, the prompt γ-rays. Results from our analysis from the spontaneous fission of ^{252}Cf and fission of ^{235}U by fast neutrons from 1 to 20 MeV are presented.

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II. EXPERIMENT

Two experiments to measure the prompt γ-ray and neutron distributions from the spontaneous fission of ^{252}Cf and neutron-induced fission of ^{235}U were carried out at in Weapons Neutron Research facility at the Los Alamos National Laboratory using the FIGARO neutron detector array [4]. The array held 17 Eljen EJ301 organic liquid scintillators, of which six were chosen for analysis. The detectors with 12.5 cm diameters by 5.0 cm depths active volumes were positioned $\sim$1 meter away from the center of the target position yielding an angular coverage of 42° to 125° relative to the neutron beam in the laboratory frame. The ^{252}Cf and ^{235}U targets, fabricated at the Lawrence Livermore National Laboratory, were housed between the cathodes and anodes of two Parallel Plate Avalanche Counters ($PPAC's$) [5]. The uranium $PPAC$,enriched to 99.91% in^{235}U, consisted of ten foils with total mass of $\sim$113 mg and the Californium $PPAC$ contained a single ^{252}Cf foil with specific activity of $\sim 2\mu Ci$. The $PPAC$'s, which served as fission tags, were designed to minimized the amount of structural material that would scatter neutrons. The fast timing of the $PPAC$'s, with $\sim$1 ns timing resolution for the photon-induced fission peak, and the EJ301 allowed for the energies of the prompt neutrons and γ-rays from fission to be resolved above $\sim$1 MeV and $\sim$120 keV, respectively. Neutrons and γ-rays events detected by the EJ301's were identified using the pulse height vs. time of flight and the pulse shape discrimination, c.f. FIG. 1 and Ref. [6], techniques. The separation between the slow (PSD1) and fast (PSD2) components of the scintillation light below around 120 keV in γ-ray energy and $\sim$1 MeV neutron energy are indistinguishable, thus both techniques were

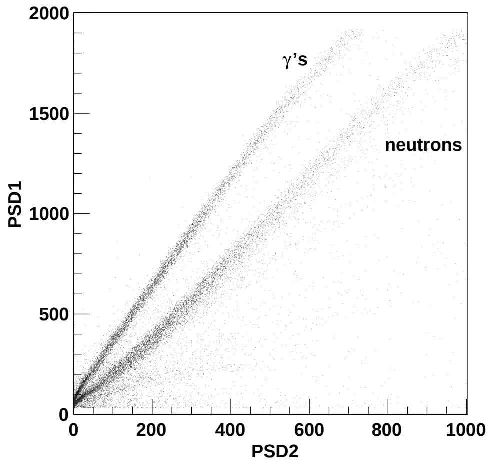

FIG. 1. The fast prompt ($PSD1$) vs. slow decay ($PSD2$) fluorescence components of the scintillation light from a EJ301 detector. Events identified as due to neutron and γ-rays interactions are labeled.

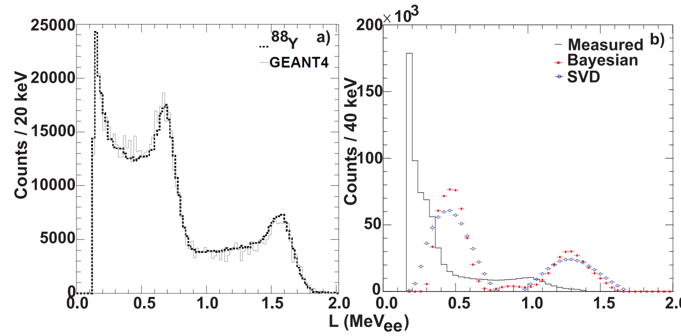

FIG. 2. (Color online) a) Comparison of GEANT4 simulation for a ^{88}Y source using known intensities in ENSDF for the 898 and 1826 keV transitions to the measured spectra. b)The unfolded spectra using SVD and the iterative Bayesian methods, open circle and filled squares, respectively, deduced from a measured spectrum of a ^{22}Na calibration source.

used to identify the interactions due to γ-ray. The chosen settings for the PSD's limited the measured energies to a range from 0.14 to ~ 4 MeV. The ability to resolve events due to neutron or γ ray interactions within the same detector is important not only because it allows us to extract information about the γ-ray and neutron spectra, but it also opens up the opportunity to study γ-neutron correlations under the same systematics while simultaneously reducing the amount of scattering material that can distort the low-energy part of the neutron spectrum. The neutrons and γ-ray distributions from fission were analyzed separately. Here, we will concentrate on testing two unfolding techniques to determine the true γ-ray distributions and the neutron spectra will be presented elsewhere.

The γ-rays spectra due to the spontaneous fission of ^{252}Cf and the neutron-induced fission at incident energies of 1-2, 5-10 and 10-20 MeV, corresponding to first, second and third chance fission, respectively, on ^{235}U were examined and compared to available data to determine whether one could unfold spectra obtained from a liquid scintillator. Due to the nature of the absorbing material in the EJ301, which consists mostly of carbon and hydrogen, the photoelectric effect tends to be suppressed compared to the Compton scattering of photons which leads to a continuum with no visible photo peak. The detector response for FIGARO was built using GEANT4 [7] to simulate the the electromagnetic interactions and determine the incident γ-ray energies. The GEANT4 model was validated using ^{22} Na, ^{60}Co and calibration sources and the SVD and iterative Bayesian unfolding techniques in the Roounfold package [8]. Shown in FIG. 2 a is a comparison of the simulation using GEANT for the 898 and 1836 keV transitions from ^{88}Y source with relative intensities of 93.7 to 99.2 [9], respectively, to the measured distribution, which includes background subtraction for natural activity in the room. The unfolding matrix obtained using GEANT4 was tested against the measured response function from calibration sources. Shown in FIG. 2b for a ^{22}Na source are the unfolded spectrum deduced from finding the eigenvalues/eigenvectors for a system of linear equations such that measured spectrum equals the sum of detector responses times the real spectrum (SVD), unfilled circles, and the spectrum determined from iteratively solving Bayes theorem (Bayesian unfolding), filled squares. The deduced ratios of the intensities for the 1274 keV transition relative to the 511 keV line are 199:100 and 188:100 from the SVD and Bayesian unfolding, respectively, slightly larger than the expected intensity of 181:100 [10]. The similarity between the Compton scattering distributions for ^{88}Y and the expected intensities for the ^{22}Na transitions was used to validate the detector efficiency predicted by our model. More details about the experimental set up and results can found in Ref. [11]. The unfolding routines in RooUnfold were tested using a measured γ-ray spectrum of the spontaneous fission of ^{252}Cf in order to investigate whether it was possible to unfold the detector response of an liquid scintillator array from a continuous energy distribution. The number of degrees of freedom used in the unfolding matrix used in the SVD method was limited to incident energies of up to 5 MeV to match the energy range of the measured spectrum. A larger response matrix with incident energies up to 6 MeV was used for the Bayesian method. The unfolded spectra using the SVD and Bayesian methods are shown in FIG. 3a. Both spectra predict a similar downward slope which drops nearly three orders of magnitude from $\sim$1.5-4 MeV and a peak around 800 keV. In the SVD spectrum, this peak is broader and washes out the structure at 1.4 MeV which was observed in the Bayesian unfolded spectrum. The Bayesian distributions is more consistent with what was observed by Ref. [12] using a 4π BaF_2 array (open circles) except below $\sim$600 keV where the detector thresholds differ. The data were normalized from $\sim$1.0-3.5 MeV for comparison purposes. Deviations from previous results taken with a NaI [13] and NE-213 detector [14] can be seen above $\sim$4 MeV, which is over

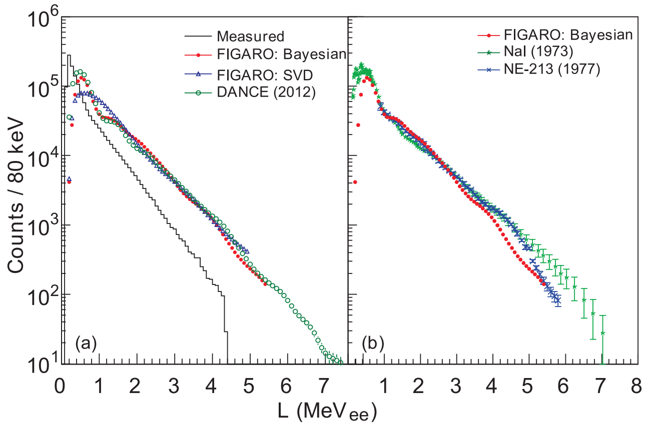

FIG. 3. (Color online) a) The unfolded ^{252}Cf spectra by the SVD (open triangles) and Bayesian (filled circles) methods from the measured spectrum (histogram) compared to the DANCE BaF_2 detectors (open circles). b) Comparison of the unfolded Bayesian spectrum with previous results obtained with NaI (open stars) [13] and NE-213 (cross) [14] detectors.

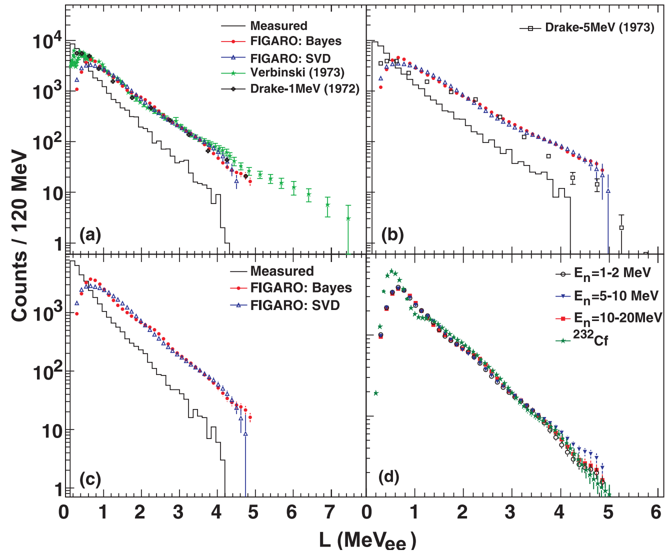

FIG. 4. (Color online) The measured and unfolded prompt γ-ray distributions using the SVD and iterative Bayesian techniques, the black, blue and red curves respectively at incident neutron energies of a)1-2 MeV, b) 5-10 MeV and c) 10-20 MeV. d) Comparison of the three curves in a) to c) deduced using the Bayesian technique and the Cf spectrum in FIG. 3.

our PSD1 range. Further measurements are need in this range to determine if the γ rays distributions decrease as rapidly as what was deduced in the current work and in Ref. [12] or has a slower drop in intensity as suggested by Ref. [13] and Ref. [14].

The γ-ray spectra from the fission of ^{235}U were also measured under similar conditions as in the ^{252}Cf experiment except the PPAC was irradiated with fast neutrons up to 100 MeV. The data was examined at three energy ranges corresponding to regions where first- (E_n =1-2 MeV), second- (5-10 MeV), and third-chance (10-20 MeV) fission, see FIG. 4a-c, respectively, are known to occur in order to determine whether the distributions change with neutron energy. All events detected by the six chosen liquid scintillator detectors were summed for statistical purposes; thus any anisotropy due to the angular distributions was neglected. The distributions were unfolded using a response matrix generated from GEANT4 for incident γ-ray energies up to 5 MeV. The incident spectra deduced from the two unfolding techniques,the iterative Bayesian and SVD are essentially identical above 1.8 MeV. A comparison of the unfolded pulse height spectra shown in FIG. 4d, indicate that the distributions below 4 MeV are insensitive to the incident neutron energy. The shape of the distributions between thermal measured by Verbinski *et al* [13] and 1 MeV neutron energies measured by Drake [15] also agree with the current measurements. The disagreement below 800 keV is due to the threshold at 130 keV in the current work and due to poor resolution of the NE-213 detectors, the oscillations below 0.7 MeV observed by Verbinski can not be seen. At ∼5 MeV neutron energy, the broad resonance at ∼2.5 MeV observed by Drake [16] is not seen in the current work.

The population of different mass fragments from fission by different materials may lead to variations in prompt γ-ray spectrum observed. Experiments using the Crystal ball array found enhancements in the γ-ray spectra at ∼4-6 MeV for nuclei around mass ∼130 [17]. Verbinski *et al.* found a systematic softening of the γ-ray spectra with increasing mass number of the fissioning isotope at photon energies above ∼3.5 MeV. Between 0.7 and ∼3.5 MeV his unfolded prompt γ-ray energy spectra for ^{235}U, ^{239}Pu and ^{252}Cf were found to be similar. A comparison of the distributions from the current work for the prompt γ rays from fission of ^{235}U and ^{252}Cf is given in FIG 4d. The distribution from the spontaneous fission of ^{252}Cf has been normalized to the distribution from the neutron-induced fission ^{235}U from 1 to 4 MeV for the purpose of comparison. The variations in near the end of the distributions arise from the differences in the measured statistics at around 4 MeV. Except for the fine details in the californium distribution, both distributions have the same monotonically decreasing trend with approximately the same slope in the energy region from1to 4 MeV. Future measurements with improved statistics will be carried out using ^{235}U and ^{239}Pu targets to measure the distributions past 4 MeV to determine if the softening at the higher γ-ray energies observed by Verbinski *et al.* is a global feature for neutron-induced fission.

III. CONCLUSION

Two experiments measuring the prompt γ-ray distributions from the spontaneous fission of ^{252}Cf and neutron-induced fission of ^{235}U were carried out at LANSCE using the FIGARO neutron array. Two unfolding techniques, SVD and iterative Bayesian, were tested to determine the fission γ-ray distributions from ^{252}Cf and from fast neutrons on ^{235}U. The techniques were validated using standard calibrations sources and with comparison with published distributions on the spontaneous fission of ^{252}Cf. The iterative Bayesian method is able to reproduce the finer details observed in other measurements while the SVD approach yields a broader distribution and smooths any fine details. The same monotonically decreasing slope was observed for the γ-ray energies from 1.0 to 4.0 MeV for ^{252}Cf and the ^{235}U regardless of incident neutron energy. The success of using the modern unfolding techniques to unfold the γ-ray distributions from fission with a liquid scintillator array will pave the way for future studies on γ-neutron correlations needed to improve the predictive capabilities for neutron and γ-ray emissions in fission models.

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[1] The Organisation of Economic Cooperation and Development's Nuclear Energy Agency, The Role of Nuclear Energy in a Low-carbon Energy Future, NEA no. 6887 (2012).
[2] Nuclear Engineering Theory and Technology of Commercial Nuclear Power, 2nd edition, Taylor & Francis, pp 46 (1992).
[3] A. Lüthi, Ph.D. Thesis, École Polytechnique Fédérale de Lausanne (1998).
[4] R. C. Haight *et al.*, in Proceedings of the Eleventh International Symposium on Capture Gamma-Ray Spectroscopy and Related Topics, edited by J. Kvasil, P. Cejnar, and M. Krticka (World Scientific Publishing Co., Singapore, 2003), pp. 451459.
[5] C. Y. Wu *et al.*, LLNL-TR-461044, 2010.
[6] G. F. Knoll, Radiation Detection and Measurement, 3rd edition, John Wiley and Sons, New York, 1994.
[7] S. Agostinelli *et al.* Nucl. Instr. and Meth. A **506**, 250 (2003).
[8] T. Adye, ArXiv:phyics.data-an/1105.1160v1.
[9] G. Mukherjee and A. A. Sonzogni, Nuclear Data Sheets **105**, 419 (2005).
[10] R. B. Firestone, Nuclear Data Sheets **106**, 106 (2005).
[11] E. Kwan *et al.*, Nucl. Instr. and Meth. A **688**, 55 (2012).
[12] A. Chyzh *et al.* Phys. Rev. C, **85**, 021601R (2012).
[13] V. V. Verbinsk *et al.*, Phys. Rev. C **7**, 1173 (1973).
[14] D. T. Ingersoll and B. W. Wehring, Nucl. Inst. and Meth. A, **147**, 551 (1977).
[15] D. M. Drake, Los Alamos Scientific Laboratory Report No. LA-5048, 1972.
[16] D. M. Drake ,Los Alamos Scientific Laboratory Report No. LA-5134, 1973.
[17] J. B. Fitzgerald *et al.*, Z. Phys. A **355**, 401 (1996).